

FORM PTO 1390 (REV 5-99) US DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE		ATTORNEY DOCKET NUMBER 2001-0462A
<b>TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. §371</b>		U.S. APPLICATION NO. <b>09/7830074</b> (if known, see 37 CFR 1.52) [NEW]
International Application No. <b>PCT/EP99/08462</b>	International Filing Date <b>November 5, 1999</b>	Priority Date Claimed <b>November 13, 1998</b>
<b>Title of Invention</b> <b>METHOD FOR PRODUCING PURE MELAMINE</b>		
<b>Applicant(s) For DO/EO/US</b> <b>Gerhard COUFAL</b>		
Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:		
<ol style="list-style-type: none"> <li>1. <input checked="" type="checkbox"/> This is a <b>FIRST</b> submission of items concerning a filing under 35 U.S.C. §371.</li> <li>2. <input type="checkbox"/> This is a <b>SECOND</b> or <b>SUBSEQUENT</b> submission of items concerning a filing under 35 U.S.C. §371.</li> <li>3. <input checked="" type="checkbox"/> This express request to begin national examination procedures (35 U.S.C. §371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. §371(b) and PCT Articles 22 and 39(1).</li> <li>4. <input checked="" type="checkbox"/> A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.</li> <li>5. <input checked="" type="checkbox"/> A copy of the International Application as filed (35 U.S.C. §371(c)(2))             <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> is transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input checked="" type="checkbox"/> has been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> is not required, as the application was filed in the United States Receiving Office (RO/US)</li> </ol> </li> <li>6. <input checked="" type="checkbox"/> A translation of the International Application into English (35 U.S.C. §371(c)(2)). <b>ATTACHMENT A</b></li> <li>7. <input type="checkbox"/> Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. §371(c)(3)).             <ol style="list-style-type: none"> <li>a. <input type="checkbox"/> are transmitted herewith (required only if not transmitted by the International Bureau).</li> <li>b. <input type="checkbox"/> have been transmitted by the International Bureau.</li> <li>c. <input type="checkbox"/> have not been made; however, the time limit for making such amendments has NOT expired.</li> <li>d. <input type="checkbox"/> have not been made and will not be made.</li> </ol> </li> <li>8. <input type="checkbox"/> A translation of the amendments to the claims under PCT Article 19.</li> <li>9. <input checked="" type="checkbox"/> An oath or declaration of the inventor(s) (35 U.S.C. §371(c)(4)). <b>ATTACHMENT B</b></li> <li>10. <input type="checkbox"/> A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. §371(c)(5)).</li> </ol>		
<b>Items 11. to 14. below concern other document(s) or information included:</b>		
<ol style="list-style-type: none"> <li>11. <input checked="" type="checkbox"/> An Information Disclosure Statement under 37 CFR 1.97 and 1.98. <b>ATTACHMENT C</b></li> <li>12. <input type="checkbox"/> An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.</li> <li>13. <input checked="" type="checkbox"/> A <b>FIRST</b> preliminary amendment. <b>ATTACHMENT D</b> <ul style="list-style-type: none"> <li><input type="checkbox"/> A <b>SECOND</b> or <b>SUBSEQUENT</b> preliminary amendment.</li> </ul> </li> <li>14. <input checked="" type="checkbox"/> Other items or information:             <ol style="list-style-type: none"> <li>a. Cover Page of Published International Application WO 00/29393 - <b>ATTACHMENT E</b></li> <li>b. International Search Report - <b>ATTACHMENT F</b></li> </ol> </li> </ol>		

U.S. APPLICATION NO. <b>09/830074</b> [NEW]	INTERNATIONAL APPLICATION NO. PCT/EP99/08462	ATTORNEY'S DOCKET NO. 2001-0462A																
<input checked="" type="checkbox"/> 15. The following fees are submitted		CALCULATIONS PTO USE ONLY																
<b>BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):</b> Neither international preliminary examination fee nor international search fee paid to USPTO and International Search Report not prepared by the EPO or JPO ..... \$1000.00 International Search Report has been prepared by the EPO or JPO ..... \$ 860.00 International preliminary examination fee not paid at USPTO but international search paid to USPTO ..... \$ 710.00 International preliminary examination fee paid to USPTO but claims did not satisfy provisions of PCT Article 33(1)-(4) ..... \$ 690.00 International preliminary examination fee paid at USPTO and all claims satisfied provisions of PCT Article 33(1)-(4) ..... \$ 100.00																		
<b>ENTER APPROPRIATE BASIC FEE AMOUNT =</b>		\$860.00																
Surcharge of \$130.00 for furnishing the oath or declaration later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(e)).																		
<table border="1" style="width: 100%; border-collapse: collapse;"> <thead> <tr> <th style="width: 25%;">Claims</th> <th style="width: 25%;">Number Filed</th> <th style="width: 25%;">Number Extra</th> <th style="width: 25%;">Rate</th> </tr> </thead> <tbody> <tr> <td>Total Claims</td> <td>11 -20 =</td> <td>0</td> <td>X \$18.00</td> </tr> <tr> <td>Independent Claims</td> <td>1 - 3 =</td> <td>0</td> <td>X \$80.00</td> </tr> <tr> <td colspan="2">Multiple dependent claim(s) (if applicable)</td> <td colspan="2">+ \$270.00</td> </tr> </tbody> </table>			Claims	Number Filed	Number Extra	Rate	Total Claims	11 -20 =	0	X \$18.00	Independent Claims	1 - 3 =	0	X \$80.00	Multiple dependent claim(s) (if applicable)		+ \$270.00	
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<b>TOTAL OF ABOVE CALCULATIONS =</b>		\$860.00																
<input type="checkbox"/> Small Entity Status is hereby asserted. Above fees are reduced by 1/2.																		
<b>SUBTOTAL =</b>		\$860.00																
Processing fee of \$130.00 for furnishing the English translation later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(f)). +																		
<b>TOTAL NATIONAL FEE =</b>		\$860.00																
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). \$40 per property +																		
<b>TOTAL FEES ENCLOSED =</b>		\$860.00																
Amount to be refunded \$ _____		Amount to be charged \$ _____																
a. <input checked="" type="checkbox"/> A check in the amount of <u>\$ 860.00</u> to cover the above fees is enclosed. A duplicate copy of this form is enclosed. b. <input type="checkbox"/> Please charge my Deposit Account No. 23-0975 in the amount of \$ _____ to cover the above fees. A duplicate copy of this sheet is enclosed. c. <input checked="" type="checkbox"/> The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. <u>23-0975</u> .																		
<b>NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.</b>																		
19. CORRESPONDENCE ADDRESS																		
 000513 PATENT TRADEMARK OFFICE		By: <u>Matthew Jacob</u> Matthew Jacob, Registration No. 25,154  WENDEROTH, LIND & PONACK, L.L.P. 2033 "K" Street, N.W., Suite 800 Washington, D.C. 20006-1021 Phone: (202) 721-8200 Fax: (202) 721-8250  April 23, 2001																

THE COMMISSIONER IS AUTHORIZED  
TO CHARGE ANY DEFICIENCY IN THE  
FEE FOR THIS PAPER TO DEPOSIT  
ACCOUNT NO. 23-0975.

[CHECK NO. UN13]  
[2001-0462A]

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of

Gerhard COUFAL : Attn: BOX PCT  
Serial No. [NEW] : Docket No. 2001-0462A  
Filed April 23, 2001 : THE COMMISSIONER IS AUTHORIZED  
METHOD FOR PRODUCING PURE TO CHARGE ANY DEFICIENCY IN THE  
MELAMINE FEE FOR THIS PAPER TO DEPOSIT  
[Corresponding to PCT/EP99/08462 ACCOUNT NO. 23-0975.  
Filed November 5, 1999]

PRELIMINARY AMENDMENT TO REDUCE PTO FILING FEES

Assistant Commissioner for Patents,  
Washington, DC 20231

Sir:

In the interest of compact prosecution and to reduce PTO filing fees, please amend the present application as follows:

IN THE CLAIMS:

Please amend claims 3 through 10 as follows:

3. (Amended) Method according to Claim 1, characterized in that the melamine obtained according to a) or b) and present as a suspension is dissolved by feeding in an aqueous ammoniacal solution, preferably a recycled mother liquor obtained in the crystallization, the solution is optionally mixed with NaOH and, if required, allowed to dwell, the dissolved ammonia is, if required, stripped, filtration is then effected and the melamine is crystallized and isolated.

4. (Amended) Method according to Claim 1, characterized in that the melamine melt is quenched in stage a) by means of recycled mother liquor obtained in the crystallization.

5. **(Amended)** Method according to Claim 1, characterized in that the melamine melt is cooled to a temperature which is from about 1 to 50°C above the melting point of the melamine, at an ammonia pressure of from about 50 to 1000 bar while feeding in ammonia.

6. **(Amended)** Method according to Claim 1, characterized in that the melamine melt is cooled to a temperature which is from about 1 to 30°C above the melting point of the melamine.

7. **(Amended)** Method according to Claim 1, characterized in that the melamine melt is cooled to a temperature which is from about 1 to 50°C above the melting point of the melamine, by passing in ammonia for from about 1 min to 10 h.

8. **(Amended)** Method according to Claim 1, characterized in that quenching is effected in stage a) at a temperature of from about 25°C to 300°C, preferably from about 50°C to 200°C and a pressure of from about 1 to 100 bar, preferably from about 1 to 50 bar.

9. **(Amended)** Method according to Claim 1, characterized in that quenching is effected in stage b) at a temperature of from about 200°C to 270°C and a pressure of from about 1 to 100 bar, preferably from about 1 to 50 bar and further cooling is then effected in the second step to about 50°C to 200°C.

10. **(Amended)** Method according to Claim 1, characterized in that melamine and urea are washed out of the off-gases of the melamine reactor by means of a urea melt which simultaneously heats up, and the urea melt is then fed to the melamine synthesis in a melamine reactor and the off-gases are fed to a urea reactor.

R E M A R K S

The above amendment is presented to eliminate multiple dependent claims, thereby reducing PTO filing fees.

Attached hereto is a marked-up version of the changes made to the claims by the current amendment. The attached pages is entitled "Version with Markings to Show Changes Made".

Favorable action on the merits is now requested.

Respectfully submitted,

Gerhard COUFAL

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April 23, 2001

## Version with Markings to Show Changes Made

3. **(Amended)** Method according to [either of Claims] Claim 1 [and 2], characterized in that the melamine obtained according to a) or b) and present as a suspension is dissolved by feeding in an aqueous ammoniacal solution, preferably a recycled mother liquor obtained in the crystallization, the solution is optionally mixed with NaOH and, if required, allowed to dwell, the dissolved ammonia is, if required, stripped, filtration is then effected and the melamine is crystallized and isolated.

4. **(Amended)** Method according to [any of Claims] Claim 1 [to 3], characterized in that the melamine melt is quenched in stage a) by means of recycled mother liquor obtained in the crystallization.

5. **(Amended)** Method according to [any of Claims] Claim 1 [to 4], characterized in that the melamine melt is cooled to a temperature which is from about 1 to 50°C above the melting point of the melamine, at an ammonia pressure of from about 50 to 1000 bar while feeding in ammonia.

6. **(Amended)** Method according to [any of Claims] Claim 1 [to 5], characterized in that the melamine melt is cooled to a temperature which is from about 1 to 30°C above the melting point of the melamine.

7. **(Amended)** Method according to [any of Claims] Claim 1 [to 6], characterized in that the melamine melt is cooled to a temperature which is from about 1 to 50°C above the melting point of the melamine, by passing in ammonia for from about 1 min to 10 h.

8. **(Amended)** Method according to [any of Claims] Claim 1 [to 7], characterized in that quenching is effected in stage a) at a temperature of from about 25°C to 300°C, preferably from

about 50°C to 200°C and a pressure of from about 1 to 100 bar, preferably from about 1 to 50 bar.

9. **(Amended)** Method according to [any of Claims] Claim 1 [to 8], characterized in that quenching is effected in stage b) at a temperature of from about 200°C to 270°C and a pressure of from about 1 to 100 bar, preferably from about 1 to 50 bar and further cooling is then effected in the second step to about 50°C to 200°C.

10. **(Amended)** Method according to [any of Claims] Claim 1 [to 9], characterized in that melamine and urea are washed out of the off-gases of the melamine reactor by means of a urea melt which simultaneously heats up, and the urea melt is then fed to the melamine synthesis in a melamine reactor and the off-gases are fed to a urea reactor.

Method for producing pure melamine

The invention relates to a method for producing pure melamine in a high-pressure process in which, 5 before solidification, the melamine melt is cooled with a supply of ammonia and is then worked up in an aqueous medium. The production of melamine by pyrolysis of urea is known, for example, from "Ullmann's Encyclopedia of Industrial Chemistry, Vol. A16, 5<sup>th</sup> ed (1990), pages 10 171-185. In the Montedison process described therein by way of example, urea is decomposed in a reactor at 370°C and 70 bar together with ammonia in the course of 20 min. The reaction mixture substantially comprising a melamine melt, ammonia and CO<sub>2</sub> is then depressurized to 15 25 bar in a quencher and is treated at 160°C with an aqueous NH<sub>3</sub>/CO<sub>2</sub> solution, solid melamine being precipitated. In order to decompose any unconverted urea or byproducts, the crude melamine suspension obtained is, if required, left in the quencher for some 20 time. Thereafter, the melamine suspension is, if required, freed from NH<sub>3</sub> and CO<sub>2</sub> in a stripper and diluted by adding mother liquor, the melamine being dissolved. After the addition of sodium hydroxide 25 solution and treatment with active carbon, the melamine is crystallized.

In a further melamine process (Nissan process), the urea decomposition is effected at 100 bar and 400°C, the urea melt employed being used before the melamine synthesis for washing melamine and urea out of 30 the off-gases of the melamine reactor. The melamine melt obtained is quenched with an aqueous ammonia solution, optionally after an ageing step, is dissolved thereby and is left to dwell at 180°C for decomposition

of impurities. After stripping of the ammonia and filtration of the solution, the melamine is crystallized. According to US 3,637,686, the melamine melt, before being quenched with aqueous ammonia, is 5 quenched in a first step with cold liquid or gaseous ammonia at a pressure of from 5 to 100 bar and a temperature from 200 to 270°C, the melamine solidifying.

The crude melamine initially obtained in the 10 melamine synthesis, which contains from about 94 to 97% by weight of melamine, depending on the production process, and in particular melam, melem, melone, ureidomelamine, ammeline and ammelide as substantial impurities, is however unsuitable or only 15 insufficiently suitable for most applications owing to the inadequate quality of the resins preparable therewith. In order to obtain a pure melamine, additional process steps, such as, for example, recrystallization, are necessary.

20 It is accordingly the object to provide a simpler method by means of which melamine can be obtained in better purity and with good yields. According to the invention, this object could be achieved if the crude melamine melt arriving from the 25 melamine reactor is cooled to just above the melting point before solidification and aqueous working-up with incorporating further ammonia.

The present invention accordingly relates to a 30 method for producing pure melamine, characterized in that the melamine melt prepared from urea in a high-pressure process, optionally after stripping of the off-gases and optionally after dwelling in an ageing container, is cooled to a temperature which is about 1

to 50°C above the melting point of melamine dependent on the respective ammonia pressure, with the addition of ammonia, after which either

- 5 a) quenching is effected with water or an aqueous ammonia- and/or melamine-containing solution or suspension and the melamine is solidified - or
- b) quenching is effected with cold liquid or gaseous ammonia, the melamine solidifying and then being further cooled in a second step with water or an aqueous ammonia- and/or melamine-containing solution or suspension and
- 10 c) the melamine is then isolated.

In melamine high-pressure processes, melamine is obtained in liquid form as a melt at pressures of 15 from about 70 to 800 bar and temperatures of at least about 370°C. The off-gases formed in the melamine synthesis and containing in particular NH<sub>3</sub> and CO<sub>2</sub> can be separated off either before or after cooling of the melt. Advantageously, the off-gases are washed by being 20 passed through a urea melt, in particular particles of melamine or of unconverted urea which are entrained by the off-gases being washed out. The urea melt is heated by the hot off-gases and is advantageously passed into a melamine reactor for melamine synthesis, while the 25 purified off-gases are advantageously passed into a urea reactor. Either the off-gases can be passed directly into the urea reactor or they are condensed, for example with the aid of ammonium carbonate or ammonium carbamate solutions which are obtained, for 30 example, in the melamine plant or the urea plant. The resulting heat can be used, for example, for preheating the ammonia used in the urea plant or for the production of steam.

After the off-gases have been separated off, the melamine melt can advantageously be stripped, for example with NH<sub>3</sub>, with the result that in particular residual CO<sub>2</sub> is removed. It is furthermore advantageous

5 to allow the melamine melt to dwell in an ageing container, as described, for example, in WO96/23778 or WO96/20182. The cooling according to the invention to the temperature of about 1 to 50°C above the melting point of melamine which is dependent on the respective

10 ammonia pressure can be effected either by means of heat exchangers or by feeding cold liquid, gaseous or supercritical NH<sub>3</sub> into the gas space above the melt or preferably by passage into the melt or by a combination of these cooling measures, thorough mixing, for example

15 by passing in NH<sub>3</sub>, possibly by additional mixing means, such as, for example, stirrers, static mixers, etc., being advantageous. Ideally, a melamine melt saturated with NH<sub>3</sub> is obtained. According to the invention, however, melamine melts supersaturated or subsaturated

20 with NH<sub>3</sub> can also be obtained at the respective pressure and the respective temperature, depending on the process conditions and on the amount of NH<sub>3</sub> fed in. The melt is preferably cooled to a temperature which is about 1 to 30°C above the melting point of the melamine

25 which is dependent on the respective ammonia pressure. It has proved particularly advantageous if cooling is effected to a temperature which is as close as possible above the melting point of melamine which is dependent on the respective ammonia pressure. Preferably, the

30 cooling is effected over a period of from about 1 min to 10 h, particularly preferably from about 1 min to 1 h. The dwell time in this temperature range is from about 1 min to 10 h, preferably from about 1 min to 1

h. The ammonia pressure during the cooling is preferably from about 50 to 1000 bar, pressures of from about 50 to 400 bar being particularly preferred. It is advantageous if a pressure increase is simultaneously 5 effected by means of the ammonia feed.

The melt can be cooled, for example, in the ageing container or in a separate suitable container or heat exchanger. The quenching carried out after the cooling of the melt is effected by mixing the cooled 10 melamine melt with water or an aqueous solution or suspension (according to a) or with ammonia and then with water or an aqueous solution or suspension (according to b). The mixing is carried out particularly advantageously and thoroughly by spraying 15 or passing the individual substances, or by spraying or passing the melt, into the initially introduced solution or suspension with simultaneous pressure reduction. For quenching of the melamine melt in stage a), particularly advantageously and economically the 20 mother liquor obtained in the solidification or crystallization and isolation of the melamine can be recycled and reused. The temperature during quenching of the melamine melt with water or with an aqueous ammonia- and/or melamine-containing solution or 25 suspension according to step a) is preferably from about 25 to 300°C, particularly preferably from about 50 to 200°C, and the pressure is from about 1 to 100 bar, particularly preferably from about 1 to 50 bar. The temperature is dependent on the operating 30 conditions in the quencher, in particular on pressure, concentrations and flow rates. The quenching according to b) with ammonia is preferably effected at from about 200 to 270°C and a pressure from about 1 to 100 bar,

particularly preferably from about 1 to 50 bar, after which, in a second cooling step, further cooling is effected with water or an aqueous ammonia- and/or melamine-containing solution or suspension, preferably 5 to about 50 to 200°C. The crystallized or solidified melamine obtained during the cooling is then isolated by separating off the mother liquor, for example by filtration or centrifuging, and is then dried, melamine being obtained in good yield and with good purity in 10 the region of about 99%, owing primarily to the omission of further purification steps.

The quenching is advantageously carried out continuously in a quench container downstream of the cooling of the melt. For example, liquid melamine 15 enters the upper part of the quench container at a temperature which is from just above the melting point, which is dependent on the respective ammonia pressure, to about 370°C and a pressure of from about 100 to 400 bar and is quenched both with water and with the 20 recycled aqueous suspension of solid melamine in an aqueous melamine solution from the quench container or the recycled mother liquor, which are likewise sprayed into the top of the quench container. The water sprayed in has, for example, a temperature of from about 25 to 25 90°C and the suspension or mother liquor sprayed in has a temperature from about 25 to 150°C at a pressure of from about 1 to 10 bar. According to this example, the temperature in the quench container is kept approximately constant, the melamine suspension in the 30 quench container is advantageously stirred, that part of the melamine suspension which is not recycled and which, in addition to solid melamine, also contains dissolved melamine and ammonia is taken off

continuously from the lower part of the quench container, the melamine is isolated by filtration or centrifuging and is dried and the mother liquor is partly recycled and partly removed.

5        In order to achieve higher and very high purities, the melamine can be recrystallized. It is also possible for the melamine obtained according to a) or b) to be dissolved, without prior isolation, directly in the suspension by feeding in an aqueous 10 ammoniacal solution, particularly preferably and economically the mother liquor obtained during this solidification or crystallization of the melamine being recycled and being used for the dissolution. Any byproducts formed by hydrolysis, especially the 15 oxyaminotriazines, such as, for example, ammeline and ammelide, can, if required, be kept in solution by adding an alkali, for example NaOH. The solution is, if required, allowed to dwell, if required dissolved NH<sub>3</sub> is also stripped off and, if required, treatment with 20 active carbon is effected. This is followed by filtration, and the melamine is crystallized, for example by further cooling and/or reduction of the pressure or application of a vacuum, and is separated from the mother liquor and dried.

25       The melamine obtained according to the present invention has a higher purity compared with conventional melamine after quenching with water, and the yield after the recrystallization is higher.

30       After the drying, the melamine can be aged for further improvement of the quality. The isolated, optionally recrystallized melamine is allowed to dwell (tempering), preferably under NH<sub>3</sub> pressure of from about 5 to about 600 bar, preferably from about 5 to 100 bar,

and at a temperature of from about 100°C to below the melting point of the melamine which is dependent on the respective NH<sub>3</sub> pressure, for a period of from about 5 min to 10 h, preferably from about 5 min to 5 h.

5 Example 1

20 g of melamine having a melam content of 2% by weight and a melem content of 1% by weight were introduced into an autoclave having a volume of 100 ml, and the autoclave was brought to a temperature of 370°C 10 with an NH<sub>3</sub> feed at an NH<sub>3</sub> pressure of 250 bar and was kept at this temperature and this pressure for 2 h. Cooling was then effected to 320°C in the course of 1 h with an NH<sub>3</sub> feed, the pressure of 250 bar being maintained, this temperature was maintained for 30 min 15 and the melamine melt was then sprayed into a second autoclave (1000 ml volume) in which an aqueous ammonia solution at a temperature of 159°C and a pressure of 12 bar was present. The melamine solidified and the temperature in the second autoclave increased to 168°C 20 and the pressure to 24 bar. After cooling of the autoclave, filtration and drying, melamine having a purity of 99.1% was obtained.

Example 2

20 g of melamine having a melam content of 2% by weight and a melem content of 1% by weight were introduced into an autoclave having a volume of 100 ml, and the autoclave was brought to a temperature of 370°C 25 with an NH<sub>3</sub> feed at an NH<sub>3</sub> pressure of 250 bar and was kept at this temperature and this pressure for 2 h. Cooling was then effected to 330°C in the course of 1 h 30 with an NH<sub>3</sub> feed, the pressure of 250 bar being maintained, this temperature was maintained for 30 min and the melamine melt was then sprayed into a second

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autoclave (1000 ml volume) in which an aqueous ammonia solution at a temperature of 62°C and a pressure of 1 bar was present. The melamine solidified and the temperature in the second autoclave increased to 81°C 5 and the pressure to 4 bar. After cooling of the autoclave, filtration and drying, melamine having a purity of 98.8% was obtained.

Example 3

20 g of melamine having a melam content of 2% 10 by weight and a melem content of 1% by weight were introduced into an autoclave having a volume of 100 ml, and the autoclave was brought to a temperature of 370°C with an NH<sub>3</sub> feed at an NH<sub>3</sub> pressure of 250 bar and was kept at this temperature and this pressure for 2 h. 15 Cooling was then effected to 350°C in the course of 1 h with an NH<sub>3</sub> feed, the pressure of 250 bar being maintained, this temperature was maintained for 30 min and the melamine melt was then sprayed into a second autoclave (1000 ml volume) in which an aqueous ammonia 20 solution at a temperature of 63°C and a pressure of 1 bar was present. The melamine solidified and the temperature in the second autoclave increased to 88°C and the pressure to 4 bar. After cooling of the autoclave, filtration and drying, melamine having a 25 purity of 98.6% was obtained.

Comparative Example 1

20 g of melamine having a melam content of 2% 30 by weight and a melem content of 1% by weight were introduced into an autoclave having a volume of 100 ml, and the autoclave was brought to a temperature of 370°C with an NH<sub>3</sub> feed at an NH<sub>3</sub> pressure of 250 bar and this temperature and this pressure was maintained for 2 h. The melamine melt was then sprayed into a second

autoclave (1000 ml volume) in which an aqueous ammonia solution having a temperature of 62°C and a pressure of 1 bar was present. The melamine solidified and the temperature in the second autoclave increased to 94°C  
5 and the pressure to 5 bar. After cooling of the autoclave, filtration and drying, melamine having a purity of 97.5% was obtained.

## Claims

1. Method for producing pure melamine, characterized in that the melamine melt prepared from 5 urea in a high-pressure process, optionally after stripping of the off-gases and optionally after dwelling in an ageing container, is cooled to a temperature which is about 1 to 50°C above the melting point of melamine dependent on the respective ammonia 10 pressure, with the addition of ammonia, after which either

- a) quenching is effected with water or an aqueous ammonia- and/or melamine-containing solution or suspension and the melamine is solidified - or
- 15 b) quenching is effected with cold liquid or gaseous ammonia, the melamine solidifying and then being further cooled in a second step with water or an aqueous ammonia- and/or melamine-containing solution or suspension and
- 20 c) the melamine is then isolated.

2. Method according to Claim 1, characterized in that the cooling of the melamine melt to the temperature which is from about 1 to 50°C above the melting point of the melamine is effected by passing in 25 cold liquid or gaseous ammonia.

3. Method according to either of Claims 1 and 2, characterized in that the melamine obtained according to a) or b) and present as a suspension is dissolved by feeding in an aqueous ammoniacal solution, preferably a 30 recycled mother liquor obtained in the crystallization, the solution is optionally mixed with NaOH and, if required, allowed to dwell, the dissolved ammonia is, if required, stripped, filtration is then effected and

the melamine is crystallized and isolated.

4. Method according to any of Claims 1 to 3, characterized in that the melamine melt is quenched in stage a) by means of recycled mother liquor obtained in 5 the crystallization.

5. Method according to any of Claims 1 to 4, characterized in that the melamine melt is cooled to a temperature which is from about 1 to 50°C above the melting point of the melamine, at an ammonia pressure 10 of from about 50 to 1000 bar while feeding in ammonia.

6. Method according to any of Claims 1 to 5, characterized in that the melamine melt is cooled to a temperature which is from about 1 to 30°C above the melting point of the melamine.

15 7. Method according to any of Claims 1 to 6, characterized in that the melamine melt is cooled to a temperature which is from about 1 to 50°C above the melting point of the melamine, by passing in ammonia for from about 1 min to 10 h.

20 8. Method according to any of Claims 1 to 7, characterized in that quenching is effected in stage a) at a temperature of from about 25°C to 300°C, preferably from about 50°C to 200°C and a pressure of from about 1 to 100 bar, preferably from about 1 to 25 50 bar.

9. Method according to any of Claims 1 to 8, characterized in that quenching is effected in stage b) at a temperature of from about 200°C to 270°C and a pressure of from about 1 to 100 bar, preferably from 30 about 1 to 50 bar and further cooling is then effected in the second step to about 50°C to 200°C.

10. Method according to any of Claims 1 to 9, characterized in that melamine and urea are washed out

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of the off-gases of the melamine reactor by means of a urea melt which simultaneously heats up, and the urea melt is then fed to the melamine synthesis in a melamine reactor and the off-gases are fed to a urea reactor.

11. Method according to Claim 10, characterized in  
that the off-gases freed from melamine and urea are  
condensed, optionally with the aid of ammonium  
carbonate solution and/or ammonium carbamate solution  
which are taken off from a urea plant or the melamine  
plant, and the resulting heat is used for preheating  
the liquid ammonia used in the urea plant or for the  
production of steam.

Leanderoff

## DECLARATION AND POWER OF ATTORNEY FOR U. S. PATENT APPLICATION

( ) Original ( ) Supplemental ( ) Substitute  PCT ( ) Design

As a below named inventor, I hereby declare that: my residence, post office address and citizenship are as stated below next to my name; that I verily believe that I am the original, first and sole inventor (if only one name is listed below) or an original, first and joint inventor (if plural inventors are named below) of the subject matter which is claimed and for which a patent is sought on the invention entitled:

Title: \_\_\_\_\_

of which is described and claimed in:

( ) the attached specification, or  
( ) the specification in the application Serial No. \_\_\_\_\_ filed \_\_\_\_\_;  
and with amendments through \_\_\_\_\_ (if applicable), or  
 the specification in International Application No. PCT/EP99/08462, filed \_\_\_\_\_, and as amended  
on \_\_\_\_\_ (if applicable).

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment(s) referred to above.

I acknowledge my duty to disclose to the Patent and Trademark Office all information known to me to be material to patentability as defined in Title 37, Code of Federal Regulations, §1.56.

I hereby claim priority benefits under Title 35, United States Code, §119 (and §172 if this application is for a Design) of any application(s) for patent or inventor's certificate listed below and have also identified below any application for patent or inventor's certificate having a filing date before that of the application on which priority is claimed:

COUNTRY	APPLICATION NO	DATE OF FILING	PRIORITY CLAIMED

I hereby claim the benefit under Title 35, United States Code §120 of any United States application(s) listed below and, insofar as the subject matter of each of the claims of this application is not disclosed in the prior United States application in the manner provided by the first paragraph of Title 35, United States Code §112, I acknowledge the duty to disclose information material to patentability as defined in Title 37, Code of Federal Regulations, §1.56 which occurred between the filing date of the prior application and the national or PCT international filing date of this application:

APPLICATION SERIAL NO.	U.S. FILING DATE	STATUS: PATENTED, PENDING, ABANDONED

ATTACHMENT B

And I hereby appoint Michael R. Davis, Reg. No. 25,134; Matthew M. Jacob, Reg. No. 25,154; Warren M. Cheek, Jr., Reg. No. 33,367; Nils Pedersen, Reg. No. 33,145; Charles R. Watts, Reg. No. 33,142; and Michael S. Huppert, Reg. No. 40,268, who together constitute the firm of WENDEROTH, LIND & PONACK, L.L.P., as well as any other attorneys and agents associated with Customer No. 000513, to prosecute this application and to transact all business in the U.S. Patent and Trademark Office connected therewith.

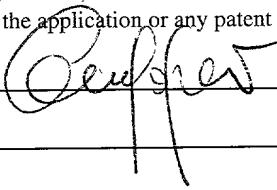
I hereby authorize the U.S. attorneys and agents named herein to accept and follow instructions from \_\_\_\_\_ as to any action to be taken in the U.S. Patent and Trademark Office regarding this application without direct communication between the U.S. attorneys and myself. In the event of a change in the persons from whom instructions may be taken, the U.S. attorneys named herein will be so notified by me.

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I further declare that all statements made herein of my own knowledge are true, and that all statements on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

1st Inventor  Date 11.4.2001

2nd Inventor \_\_\_\_\_ Date \_\_\_\_\_

3rd Inventor \_\_\_\_\_ Date \_\_\_\_\_

4th Inventor \_\_\_\_\_ Date \_\_\_\_\_

5th Inventor \_\_\_\_\_ Date \_\_\_\_\_

6th Inventor \_\_\_\_\_ Date \_\_\_\_\_

7th Inventor \_\_\_\_\_ Date \_\_\_\_\_

The above application may be more particularly identified as follows:

U.S. Application Serial No. \_\_\_\_\_ Filing Date \_\_\_\_\_

Applicant Reference Number \_\_\_\_\_ Atty Docket No. \_\_\_\_\_

Title of Invention \_\_\_\_\_